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Research Report

OPTICAL GENERATION AND DETECTION OF ACOUSTIC PULSE PROFILES IN GASES FOR NOVEL ULTRASONIC ABSORPTION SPECTROSCOPY

A. C. Tam
W. P. Leung

IBM Research Laboratory
San Jose, California 95193

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We demonstrate a new all-optical, frequency-multiplexed and fast technique for gas-phase ultrasonic absorption spectroscopy. A pulsed laser produces a short-duration acoustic pulse in the gas sample, and the acoustic pulse profiles at two distances are monitored by focused continuous probe beams. Fourier decompositions of these probe deflection signals provide the absorption spectrum, and examples for CO_2 and CO_2+H_2O mixture are given. Ultrasonic propagation speeds in the MHz regime are also obtained for several gases.

OPTICAL GENERATION AND DETECTION OF ACOUSTIC PULSE PROFILES IN GASES FOR NOVEL ULTRASONIC ABSORPTION SPECTROSCOPY

A. C. Tam
W. P. Leung[*]

IBM Research Laboratory
San Jose, California 95193

ABSTRACT: We demonstrate a new all-optical, frequency-multiplexed and fast technique for gas-phase ultrasonic absorption spectroscopy. A pulsed laser produces a short-duration acoustic pulse in the gas sample, and the acoustic pulse profiles at two distances are monitored by focused continuous probe beams. Fourier decompositions of these probe deflection signals provide the absorption spectrum, and examples for CO_2 and CO_2+H_2O mixture are given. Ultrasonic propagation speeds in the MHz regime are also obtained for several gases.

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[*]Present and Permanent Address: Department of Physics, Chinese University of Hong Kong, Shatin, N.T., Hong Kong

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Conventional acoustic absorption spectroscopy[1-5] in gases has been performed using transducers such as quartz plates, microphones, *etc.* The use of transducers for gases causes several limitations. The frequency response is usually limited to ≤ 1 MHz. Also, the frequencies are scanned point by point, and is thus slow and unsuitable for transient conditions. Short-pulsed measurements are difficult because of transducer ringing. Furthermore, transducers cannot be used in hostile environments like flames. We describe here a new all-optical, pulsed and multiplexed technique that avoids the above limitations, and is useful for acoustic absorption spectroscopy of gases. This technique relies on the use of a short-duration laser pulse to generate reliably a narrow acoustic pulse containing a broad Fourier frequency spectrum; as this pulse propagates, the various Fourier components are absorbed differently, resulting in pulse distortion that is probed by a focused CW laser beam. Fast Fourier transform of the transient probe deflection signal provides the acoustic absorption spectrum; this is much faster than the conventional point-wise frequency measurement. Our technique can be called "Opto-acoustic spectroscopy of the second kind" (OAS II) because it exploits opto-acoustic pulse generation[6-8] for *acoustic* spectroscopy. This should be distinguished from the well known opto-acoustic spectroscopy[9] (understood to be the first kind), which exploits opto-acoustic pulse generation for *optical* spectroscopy.

Our experimental demonstration of OAS II is shown in Fig. 1. A pulsed laser is used to generate a short-duration acoustic pulse in the gas under study. This is commonly called a pulsed opto-acoustic (or photo-acoustic) generation[6] which can be achieved via many mechanisms, including simple optical absorptions and nonradiative decay,[9] optical breakdown[10] and absorption at a solid surface with subsequent heat

flow to the fluid in contact.[11] The experimental arrangement in Fig. 1 uses the last

mechanism, which does not require powerful or tunable laser source, but requires an

opaque target surface to be placed in contact with the gas. Here, we use a nitrogen

laser beam at 337 nm with 1 mJ energy and 8 nsec duration as the pulsed excitation

source. The target is a polished silicon wafer which does not absorb the gases under

study, and the optically flat surface produces a plane acoustic wavefront due to the

pulsed laser heating of the Si surface. The laser spot size at the silicon surface is

about 4 mm, which is much 'arger than the acoustic wavelengths (~0.3 mm at

1 MHz) in the gas presently studied, and diffraction effect of the acoustic pulse is

small. The profile and the propagation velocity of the acoustic pulse can be detected

with short rise-time and high spatial resolution by a tightly focused probe beam[7]

which is oriented parallel to the silicon surface (at separation x). In this way, the

whole acoustic wavefront arrives simultaneously at the focused probe beam, causing a

transient angular deflection $\phi(t)$ which is uniform across the probe cross section if the

spatial extent of the acoustic pulse is much larger than the focused probe diameter

(true for our present experiment). The probe deflection $\phi(t)$ is linearly converted

into an intensity variation signal $S(x,t)$ by a fast photodiode of small active area

located where the probe beam cross-sectional area is much larger. The photodiode and

preamp assembly we used (Analogy Modules Model LNVA-O-S-100 MHz) has a

bandwidth of 100 MHz. The output signal $S(x,t)$ is further amplified by a Tektronix

7854 scope with a 7A24 plug-in (which also provides a single-shot display of $S(x,t)$),

and then accumulated on a transient recorder (Data Precision Model 6000 with Model

620 plug-in of bandwidth 30 MHz). Typically, $S(x,t)$ can be digitized for a single

laser shot, or averaged for several laser shots for signal/noise improvement. A

built-in fast Fourier transform feature in the transient recorder allows us to obtain the

Fourier spectrum $\tilde{S}(x,\omega)$ of the probe beam deflection signal $S(x,t)$, where ω is an angular frequency. The above measurement is repeated with the separation between the probe beam and the silicon target changed (by a micrometer adjustment) from x to x', obtaining another Fourier spectrum $\tilde{S}(x',\omega)$ corresponding to the probe beam deflection signal $S(x',t)$. This stepwise measurement at two separations is presently used to derive the ultrasonic absorption spectrum. A much faster measurement is possible by using two probe beams[12] at separations x and x' from the target surface for single-shot detection of the deflections.

As described in a recent opto-acoustic pulse profile study in liquids,[7] the present probe-beam deflection signal $S(x,t)$ is related to the local acoustic pressure $P(x,t)$ (with Fourier transform $\tilde{P}(x,\omega)$) by

$$S(x,t) = K\ \partial P(x,t)/\partial t \tag{1}$$

where K is a constant depending on geometry, physical properties of the gas and detection sensitivity. Fourier transform of Eq. (1) yields

$$\tilde{S}(x,\omega) = Ki\omega\ \tilde{P}(x,\omega) \tag{2}$$

where i is the imaginary unit. The pressure amplitude $\tilde{P}(x,\omega)$ propagates with a frequency-dependent absorption coefficient $\alpha(\omega)$ and velocity $v(\omega)$, and so we write

$$\tilde{P}(x,\omega) = \tilde{P}(0,\omega)\ e^{-\alpha(\omega)x+i\omega x/v(\omega)} . \tag{3}$$

Combining Eqs. (1)-(3), we have

$$\frac{\partial \ln[\mathrm{Mag}\,\tilde{S}(x,\omega)]}{\partial x} = -\alpha(\omega) \tag{4}$$

where $\mathrm{Mag}(\tilde{S}(x,\omega))$ is the magnitude of the Fourier transform of the observed probe deflection signal.

The signal $S(x,t)$ is as given by Eq. (1) only for a sufficiently small size of the detector in the direction of beam deflection, e.g., this size should be $\le 10\%$ of the diameter of the probe beam at the detector position. In such case, the magnitude and shape of the signal $S(x,t)$ critically depends on the exact position of the detector; $S(x,t)$ actually changes phase if the detector is moved along the direction of the beam deflection, and is nearly zero when it is located at the center of the probe beam cross-section, as shown in Fig. 2. Most of our experimental data are taken with the detector suitably located at a wing of the probe beam to maximize signal amplitude.

The delay time t_d with respect to the firing of the laser indicated in Fig. 2 is measured for several path lengths x for several gases at 22°C. We find that x changes linearly with t_d, and the gradient is the velocity v at the mean Fourier frequency f_m of the acoustic pulse. Some results are given in Table I. For the gases studied at 1 atmosphere and 22°C (Ar, N_2, O_2, CO_2, CF_2Cl_2, (i.e., Freon 12) and their mixture with 20 Torr of water vapor), f_m is ~1 MHz, which is larger than the dispersive frequencies in the gases studied (except mixture of CO_2 or Freon with H_2O), so that the data in Table I are the ultrasonic velocities in the "high frequency limits."

While the observed signals $S(x,t)$ for Ar, N_2, O_2 or their mixtures with water vapor do not change significantly when x is changed by ~1 mm, we observe that $S(x,t)$ for the CO_2+20 Torr H_2O mixture changes drastically with x, as shown in Fig. 3. As given by Eq. (4), the Fourier transforms of the signals exemplified in Fig. 4 for at least two displacements x provide the ultrasonic absorption spectrum $\alpha(\omega)$. Some

results for CO_2 and CO_2+20 Torr H_2O mixture at 22°C and 1 atm pressure are given in Fig. 5. As done traditionally, the vertical axis is absorption per wavelength, *i.e.*, the product of $\alpha(\omega)$ and the wavelength 2π v/ω. Our results in Fig. 5 represent the first ultrasonic absorption spectra obtained all optically in a Fourier-multiplexed manner. Previous results were obtained by point-wise frequency measurement with the use of transducers for generation and detection; the long risetimes of most transducers is a reason why high frequency data are lacking in Fig. 5. It is already well known[1,2] that "relaxational absorption" of sound at frequency ω occurs for an inelastic collisional rate on the order of ω; this is probably the reason for the large absorption feature for the CO_2 and H_2O mixture in the range of ~1 to 5 MHz. The inelastic cross-section for H_2O and CO_2 collision is unusually large (close to geometrical cross-section), and the inelastic collisional rate for the CO_2/H_2O mixture studied is ~10^7 per sec. Such relaxational ultrasonic absorption at lower H_2O concentration (corresponding to lower frequency absorption) has been observed previously.[13,14,20]

Table 1

Some Ultrasonic Velocities in Gases at 1 atm and 22°C,
Measured by Optical Acoustic Pulse Generation and Detection

Gas	Pure Gas		Gas + 20 Torr H_2O	
	Mean Frequency (MHz)	Acoustic Velocity (10^4 cm/s)	Mean Frequency (MHz)	Acoustic Velocity (10^4 cm/s)
Ar	2.5	3.218(3)	1.5	3.240(8)
N_2	2.5	3.512(5)	1.5	3.526(6)
O_2	2.5	3.295(5)	1.5	3.312(5)
CF_2Cl_2	0.7	1.504(10)	0.7	1.524(10)
CO_2	2.5	2.796(3)	1.0	2.752(6)

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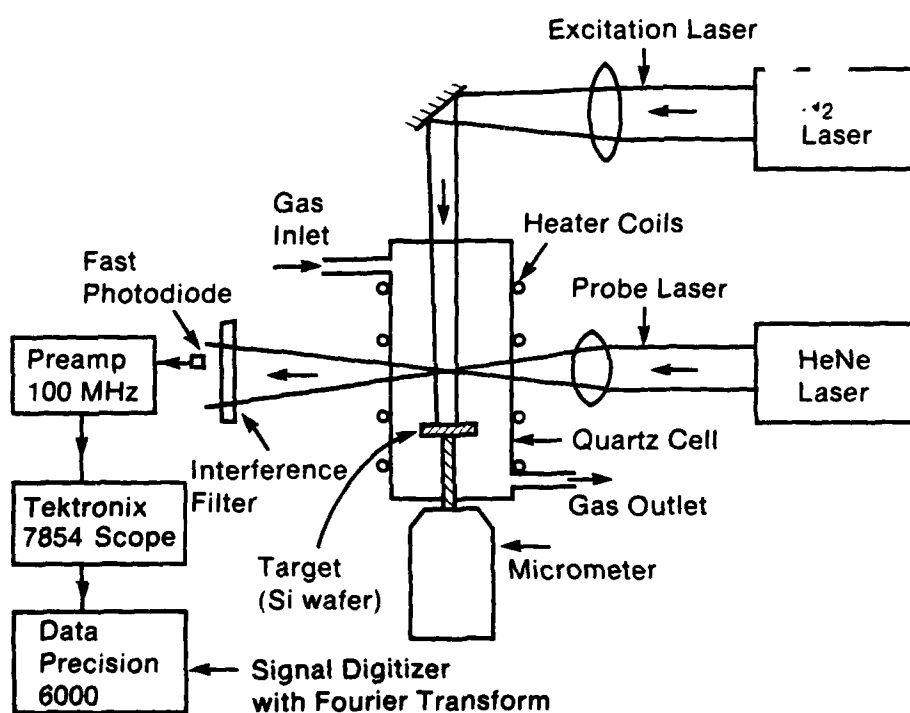
Figure 1. Experimental arrangement for the new opto-acoustic spectroscopy of the second kind. The probe deflection signal is Fourier analyzed, and the difference between two Fourier spectra at different displacements x provide the ultrasonic absorption spectrum.
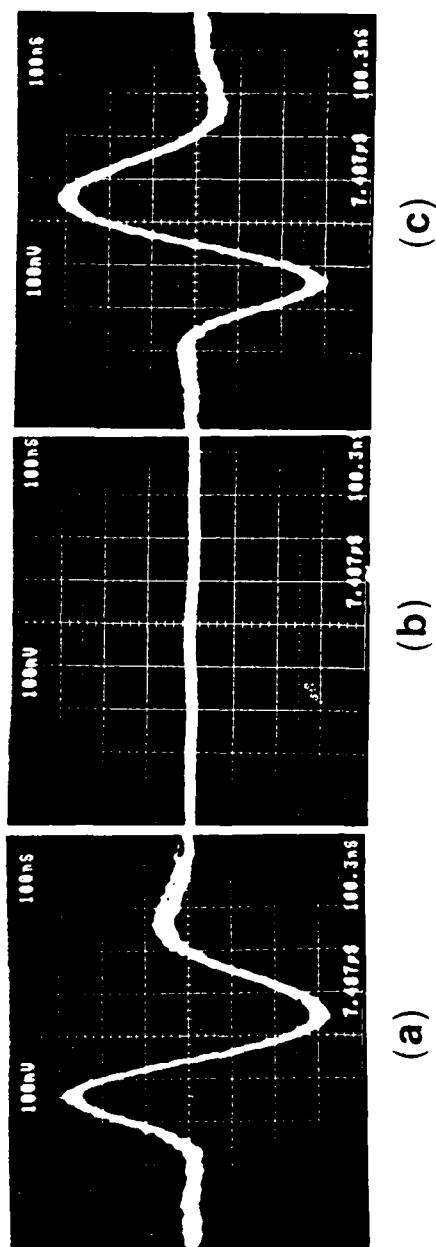
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Figure 2. Observed probe deflection signal $S(x,t)$ for dry N_2 at $22°C$ and $x = 2.7$ mm. The photo-diode is located at one wing, at center and at the other wing of the probe beam cross-section for (a), (b) and (c), respectively. Each scope picture is delayed by 7.487 μs from the laser pulse, and horizontal scale is 100 ns/division.
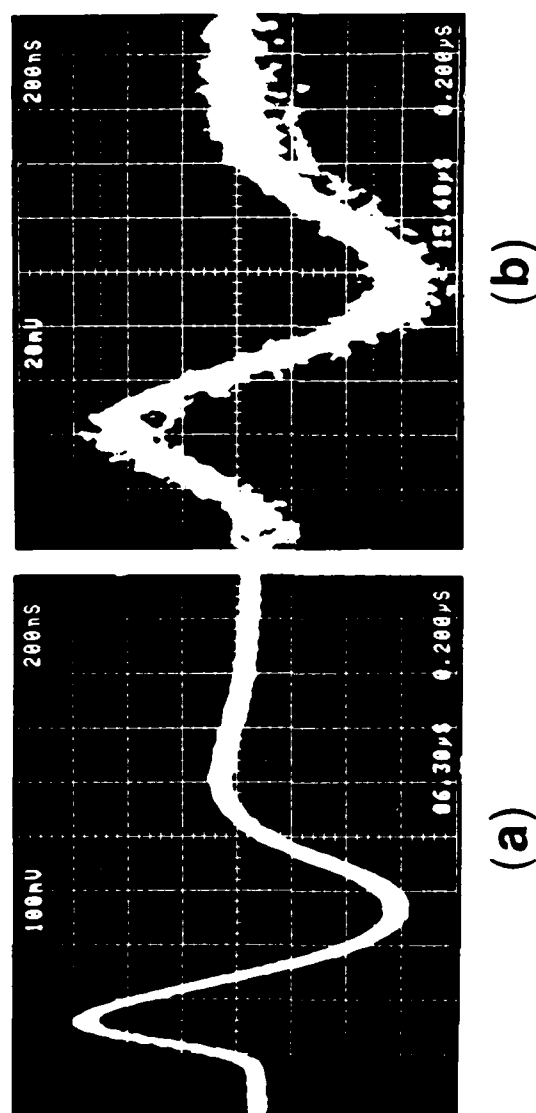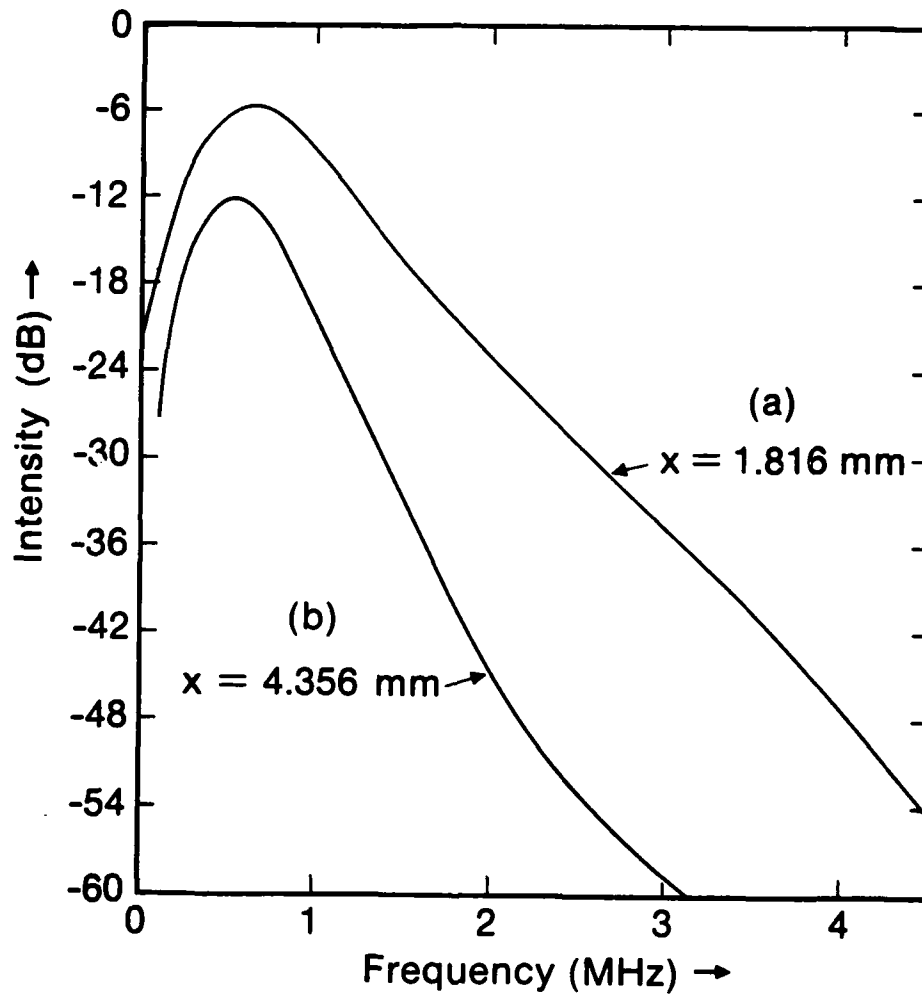
Figure 3. Observed probe deflection signal $S(x,t)$ for $CO_2 + 20$ Torr H_2O at 1 atm total pressure and 22°C. In (a), $x = 1.816$ mm and scope delay time is 6.30 μs; in (b), $x = 4.356$ mm and scope delay time is 15.40 μs. Horizontal scale is 200 ns/division for both.

Figure 4. Magnitude of the fourier transform of signals in Fig. 3 as provided by the Data Precision 6000.
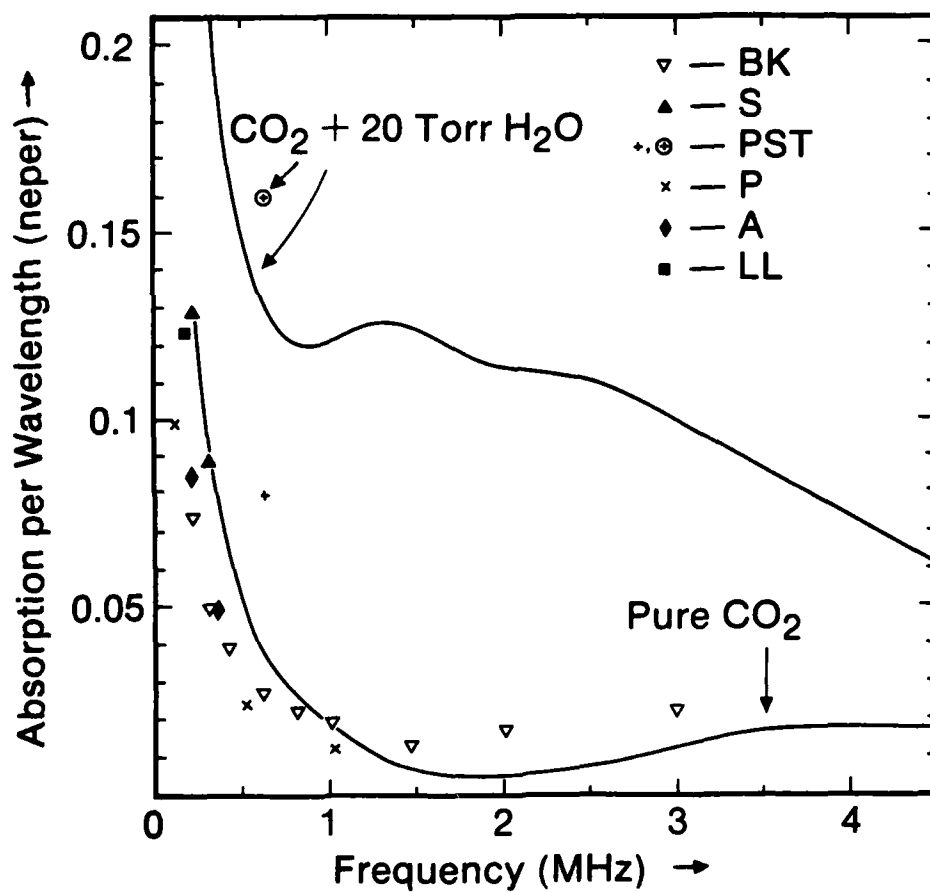
Figure 5. Ultrasonic absorption spectra derived by the use of Eq. (4) for pure CO_2, and for $CO_2 + 20$ Torr H_2O at $22°C$ and 1 atm total pressure. Previous data for comparison are: BK (Ref. 14), S (Ref. 15), PST (Ref. 16), P (Ref. 17), A (Ref. 18), LL (Ref. 19). Most of these previous data are for pure or nearly pure CO_2, except for one indicated data point.

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